

## Enhanced marine CH<sub>4</sub> emissions to the atmosphere off Oregon caused by coastal upwelling

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[1] Methane in surface waters and marine air off Oregon (44°24'N–44°54'N, 124°36'W–125°24'W) was continuously surveyed in July 1999. During a high-resolution survey after a period of steady winds from the north, CH<sub>4</sub> concentrations were high in the northeastern region, near the shelf edge. The highest CH<sub>4</sub> concentrations were 2.5 times higher than equilibrium with the atmospheric partial pressure. In contrast, concentrations were near equilibrium in the western part of the survey area, the Hydrate Ridge. The increase in CH<sub>4</sub> from southwest to northeast correlates with a drop in sea surface temperature (SST), from 16.5°C to <13.5°C, toward the shelf edge. The observed SST pattern was caused by summer upwelling off Oregon. The results suggest that CH<sub>4</sub> derived from bottom sources near the shelf/slope break and methane found in connection with shallow (100–300 m) turbidity layers is transported to the surface by coastal upwelling, which causes an enhanced net flux of CH<sub>4</sub> to the atmosphere. Vertical profiles of the methane distribution on the shelf in October demonstrate the accumulation of methane introduced by shelf sources. Surface concentrations at these stations in October (during nonupwelling conditions) were lower than in July (during upwelling) and were only slightly oversaturated with respect to the atmosphere. An acoustic Doppler current profiler survey indicates that the observed trend cannot be attributed to a surface flow reversal in the area. The low-salinity waters in the core of the Columbia River plume (*S* < 31) showed no enhanced CH<sub>4</sub> concentrations. The trend of higher CH<sub>4</sub> concentrations at lower temperatures existed over the whole 17-day survey, but large spatial and temporal variations existed. The presence of methane sources in regions of coastal upwelling worldwide, such as shallow seeps, gas hydrates, and intermediate nepheloid layers, suggests that the enhancement of CH<sub>4</sub> fluxes to the atmosphere by coastal upwelling occurs on a global scale.

**INDEX TERMS:** 1050 Geochemistry: Marine geochemistry (4835, 4850); 4279 Oceanography: General: Upwelling and convergences; 4504 Oceanography: Physical: Air/sea interactions (0312); 4820 Oceanography: Biological and Chemical: Gases; 9355 Information Related to Geographic Region: Pacific Ocean; **KEYWORDS:** upwelling, methane, air-sea flux, gas seepage

### 1. Introduction

[2] The atmospheric content of methane influences the Earth's radiation budget because of the absorption in the infrared spectrum and also because of photochemical reactions in the atmosphere [Lashof and Ahuja, 1990; Lelieveld *et al.*, 1993]. Airborne methane has more than doubled during the last 150 years because of human activities, accounting for ~22% of the “anthropogenic greenhouse effect” [Lelieveld *et al.*, 1998]. The overall contribution of

the marine environment as a source of atmospheric methane, although still uncertain because of the high variability in estuarine and shelf waters, was estimated to be ~2% of the total [Bange *et al.*, 1994; Bates *et al.*, 1996; Cicerone and Oremland, 1988]. Most of the marine sea-air methane flux occurs at the shelf and ocean margin [Bange *et al.*, 1994; Rehder and Suess, 2001]. It is generally agreed that the oceans play a minor role in today's atmospheric CH<sub>4</sub> budget.

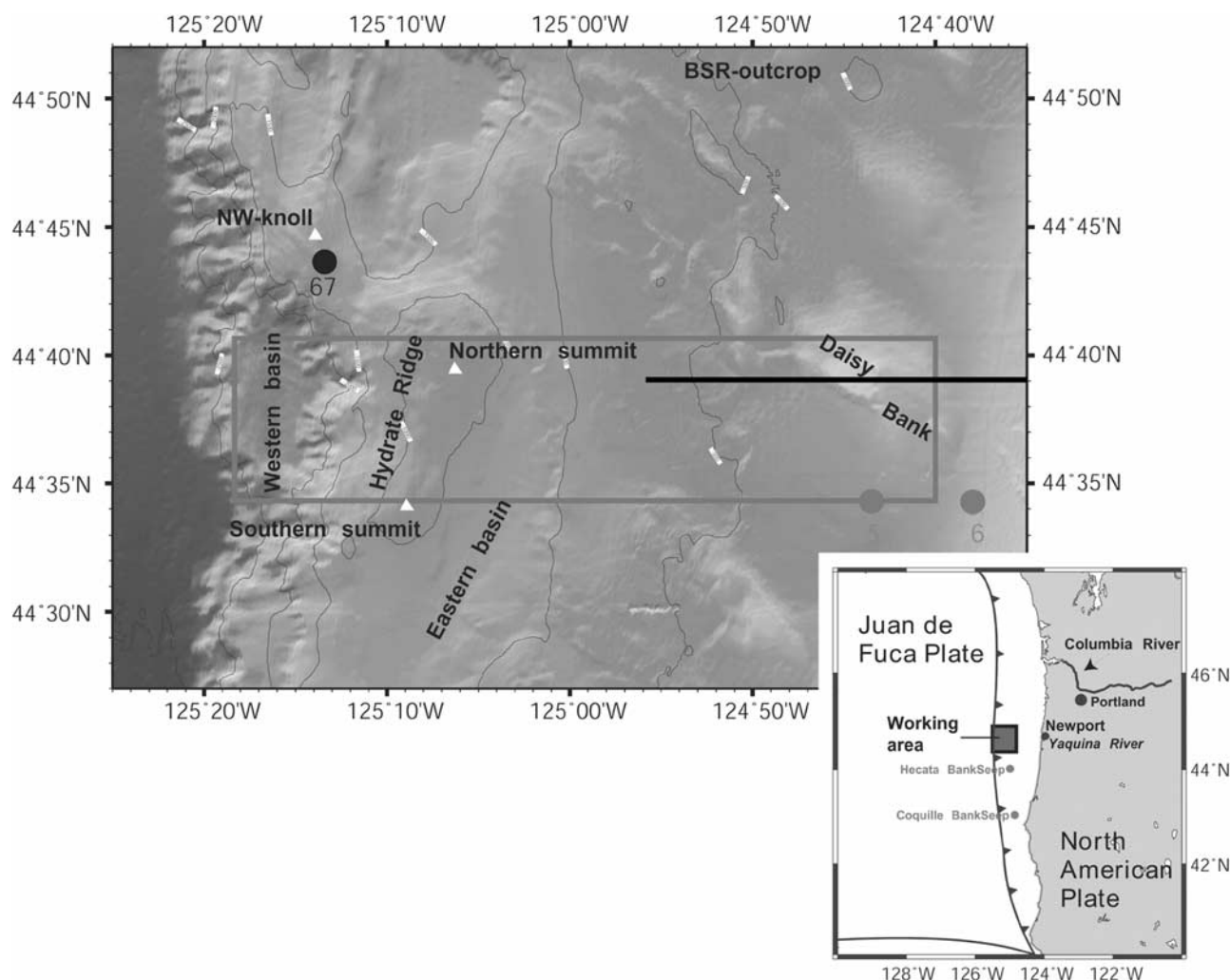
[3] While continental margins are potentially the most significant sources of CH<sub>4</sub> to the oceans, their complexity challenges simple description. The release of methane or methane-bearing fluids from the seafloor along active subduction zones has been reported in many locations along the continental margins [Moore and Vrolijk, 1992; Suess *et al.*, 1998]. An impact on the upper water column and the atmosphere, however, is not observed in most cases. Most of the known marine CH<sub>4</sub> hydrate reservoirs are also located along the continental margin [Gornitz and

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**Figure 1.** Map of working area on the continental margin off Oregon. Red rectangle denotes region of surface survey from 24 July shown in Figures 2a–2d. Black line is western part of track of current, temperature, and salinity section from same day shown in Figures 3a and 3b. Red dots indicate hydrocast stations from October 1999, shown in Figure 8. Black dot shows location of Station 67, chosen to illustrate occurrence of midwater CH<sub>4</sub> maxima in connection with intermediate nepheloid layers (Figure 9). More locations specified in the text, as well as the subduction front, are indicated in overview map (lower right). See color version of this figure at back of this issue.

Fung, 1994; Kvenvolden, 1998]. Our observations presented here were made at the Cascadia margin off Oregon (Figure 1), one of the areas studied most intensely for these accretionary processes. In this area, methane is injected into the water column during tectonically induced dewatering [Carson *et al.*, 1990]. Large near-bottom gas hydrate reservoirs have been located at 600–800 m depth, and active gas vents were detected in the vicinity of these hydrates [Suess *et al.*, 1999]. In addition, methane sources of both biogenic and thermogenic origin have been located on the nearby shelf.

[4] This work focuses on an enhancement of the methane flux to the atmosphere by coastal upwelling. This hydrographic process that occurs along large parts of the world's ocean boundaries can shortcut the pathway of methane

introduced in subsurface and intermediate waters to the surface ocean and the atmosphere.

## 2. Methods and Field Work

[5] Most data were gathered during expedition 143-1b of R/V *Sonne* from 15 to 28 July 1999 at the continental margin off Oregon (Figure 1). Surface temperature and salinity were recorded by the ship's thermosalinograph. Concentrations of methane in surface seawater and marine air were determined using a continuously running equilibration system based on gas chromatography [Rehder *et al.*, 1999; Rehder and Suess, 2001]. The inlet of the pumping system was installed in the moonpool of the ship at approximately the same depth as the thermosalinograph

inlet (6 m). Although the system was running continuously during the cruise, the ship was mostly stationary during geological fieldwork at the second accretionary ridge (Hydrate Ridge). After elimination of some data, which were contaminated by shipboard operations, 906 data points remained from the continuous survey. We will focus on a subset of these samples, collected during a 10-hour high-resolution survey performed on 24 July, in order to image the larger scale methane surface distribution (112 data points). At that time, the system was run manually to provide higher sampling frequency. Sampling was performed by extracting a 1-mL gas sample from the equilibration chamber through a septum port and injecting into the gas chromatograph (GC) every 4 min. This nonautomated use of the system is described by *Rehder et al.* [1998]. The GC was calibrated every hour, using mixtures of  $1.936 \pm 0.003$  ppmv and  $9.854 \pm 0.006$  ppmv CH<sub>4</sub> in synthetic air (Deuste Steininger GmbH, calibrated against National Oceanic and Atmospheric Administration/Climate Monitoring and Diagnostics Laboratory standards at the Institute for Environmental Physics, Heidelberg, Germany). The difference between temperatures in situ and in the equilibration chamber was accounted for by calculating the change in solubility [*Wanninkhof*, 1992] as described by *Rehder and Suess* [2001]. The saturation of methane relative to equilibrium with the atmosphere was calculated with the relation

$$\text{Saturation} = 100(c_w/c_{\text{air}}),$$

where  $c_w$  is the measured concentration in seawater and  $c_{\text{air}}$  is the concentration that would be in equilibrium with the ambient methane partial pressure of the atmosphere. The latter was calculated from the in situ atmospheric total pressure recorded by the ship's meteorological station and from the mean value of the molar fraction of methane from the continuous air measurements during the entire cruise, 1.780 ppmv. This value represents a mean of 396 measurements ( $\sigma = 0.017$ ). The mixing ratio of methane showed no long-term trend during the 14-day survey, which was the decisive factor for using the mean value for the calculation of the saturation state of seawater. A detailed description of the analytical system used as well as the calculations involved is given by *Rehder and Suess* [2001]. The methane profiles presented here were obtained using the modified vacuum degassing method [*Lammers and Suess*, 1994; *Schmitt et al.*, 1991] as described by *Rehder et al.* [1999]. The precision of the automated equilibration system, determined during station work in homogenous open ocean surface waters with a background CH<sub>4</sub> concentration in equilibrium with the atmosphere in the North Atlantic, is better than 1%. Considering the error of the calibration gases and temperature recording, we estimate the accuracy of the continuous system to be <2%. The manual use of the system, while following the same principle, adds the error caused by the use of a gas-tight syringe relative to a temperature-stabilized sample loop, leading to a total accuracy of 3.5%. Comparison of the measurements from the end of the manual survey and the restart of the automated mode (immediately afterward at the same location; data not shown) do not indicate any offset between the two data

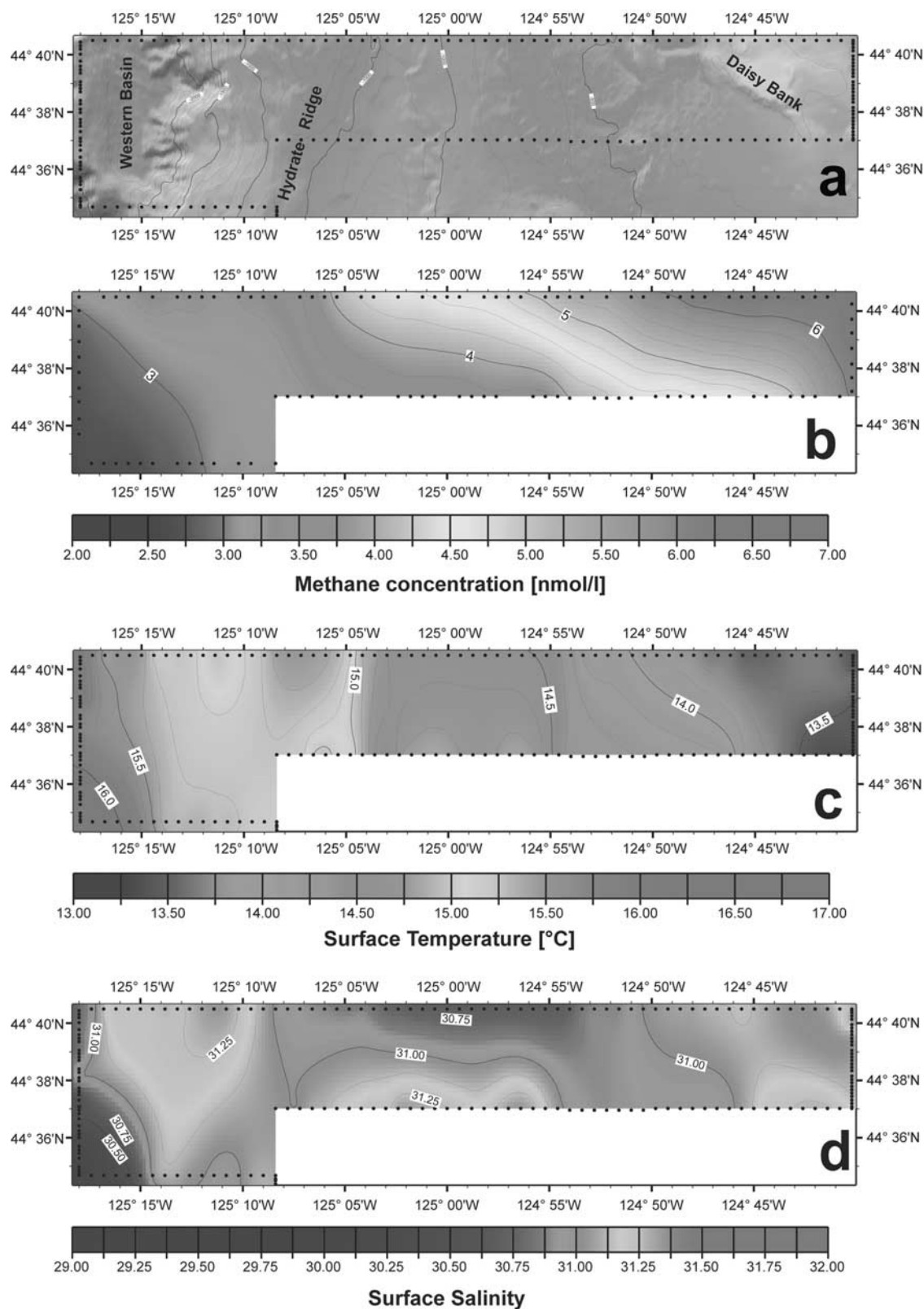
sets. The average precision of the vacuum degassing method used for discrete water samples has been determined to be ~4%, based on replicate measurements [*Rehder et al.*, 1999]. In the same work, it has also been shown that the vacuum degassing method yields, at least for surface water samples, the same absolute concentration values as the well-established continuous equilibration technique. We are therefore confident that the methane concentrations measured with these three different techniques are consistent and directly comparable.

[6] The temperature and along-slope current sections shown in Figures 3a and 3b in section 3 were obtained on R/V *Wecoma* on the same day (24 July) as the high-resolution surface CH<sub>4</sub> survey. For the temperature profile across the shelf, a towed, undulating vehicle, SeaSoar [*Pollard*, 1986], equipped with a Sea-Bird 9/11 plus conductivity-temperature-depth unit [*Barth et al.*, 2000] was used. The current velocity profile was obtained using a hull-mounted 153.6-kHz narrow-band acoustic Doppler current profiler (RD Instruments) and was processed as described by *Kosro* [2002].

### 3. Results and Discussion

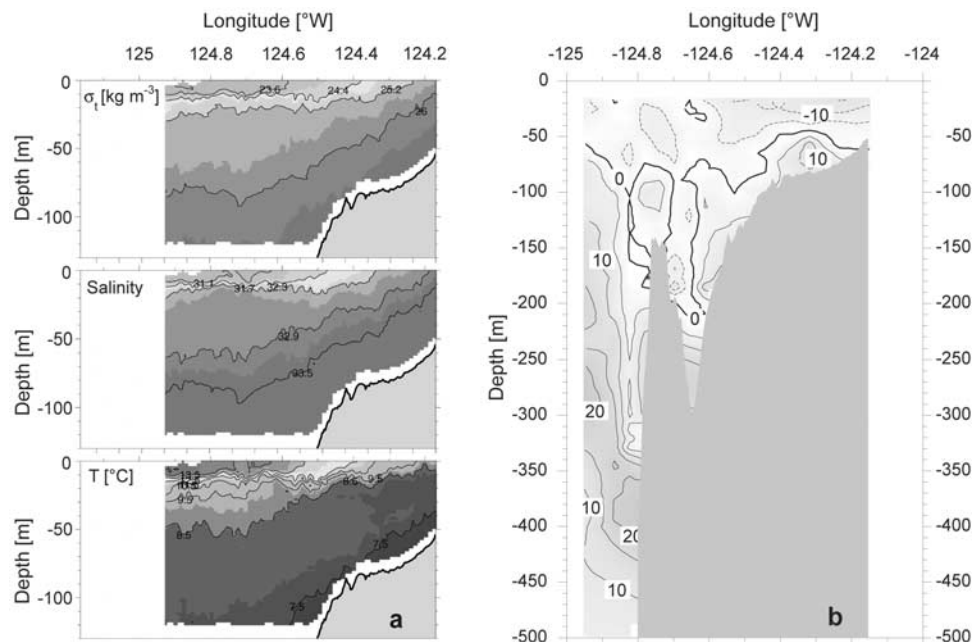
[7] During the high-resolution survey on 24 July 1999, CH<sub>4</sub> surface concentrations across the continental slope off Newport were measured over seafloor depths of 3000–300 m, including parts of the Western Basin, the second accretionary ridge (Hydrate Ridge), and the shelf near Daisy Bank (Figure 2a). The CH<sub>4</sub> surface concentrations varied from near equilibrium with the atmosphere ( $2.65 \text{ nmol L}^{-1}$ ) in the west to >250% saturation ( $6.4 \text{ nmol L}^{-1}$ ) in the northeast (Figure 2b). The gradient suggests even higher concentrations toward the shore. In the following, it will be shown that the most likely scenario to explain this pattern is upwelling of CH<sub>4</sub>-rich subsurface waters.

[8] It is well known that riverine runoff generally has methane contents significantly above those of open ocean waters [*de Angelis and Lilley*, 1987; *Scranton and McShane*, 1991]. With the Columbia River discharging ~180 km to the north and the Yaquina estuary discharging to the east [*Butler et al.*, 1987], the potential influence of riverine input on the methane distribution has to be considered. However, evidence for an inverse correlation between CH<sub>4</sub> concentrations (Figure 2b) and surface salinity (Figure 2d), which would indicate a freshwater origin of the gas, was not found. The lowest salinities within the survey area were observed in the southwest, where CH<sub>4</sub> concentrations were lowest. The decreasing surface salinity toward the west is consistent with earlier findings suggesting that the core of the Columbia River discharge can be >100 km offshore in summer [*Huyer*, 1977]. Hence it can be ruled out that the enhanced methane concentrations result from the influence of methane-rich freshwater released from the Columbia River. Approaching Astoria, Oregon, on 29 July 1999, we were able to determine the methane content of the pristine Columbia River discharge in the estuary near Astoria to be ~215 nmol L<sup>-1</sup>, or ~80 times supersaturated (data not shown). This is in agreement with a concentration of 241–279 nmol L<sup>-1</sup> reported by *Lilley et al.* [1996] within this region in October



**Figure 2.** Results from a high-resolution CH<sub>4</sub> surface survey from 24 July 1999 showing (a) bathymetry; (b) surface methane concentration, (c) surface seawater temperature, and (d) surface seawater salinity. Black dots indicate locations of individual measurements. See color version of this figure at back of this issue.





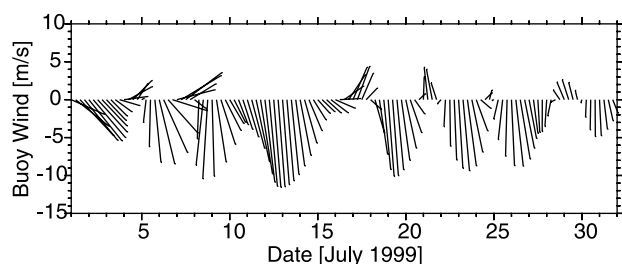
**Figure 3.** (a) The  $\sigma_t$ , salinity, and temperature sections from a SeaSoar survey and (b) section of alongshore current (in  $\text{cm s}^{-1}$ , positive northward) derived from an acoustic Doppler current profiler survey. Both data sets along  $44^\circ 39' \text{N}$  off Newport were recorded on 24 July 1999 on R/V *Wecoma* (see Figure 1 for location, but note that the section extends 30 km farther east than shown in Figure 1). Flow pattern suggests that observed temperature and CH<sub>4</sub> variations do not result from different surface current regimes. *SeaSoar* data indicate the presence of upwelled waters at the sea surface during that time. See color version of this figure at back of this issue.

1988 and with a concentration of  $128 \text{ nmol L}^{-1}$  at a salinity of 7 reported for summer 1995 [Sansone *et al.*, 1999]. The latter study also showed that the Columbia River still influences the surface ocean methane content and isotopic composition 12 km to the west of its mouth but does not affect the surface methane concentration 200 km offshore. The low methane content in the low-salinity waters in the western part of the survey presented here suggests that the Columbia River water also completely loses its high methane content on its way 180 km down the coast to  $44^\circ 40' \text{N}$ . The outflow of Yaquina Bay,  $\sim 30$  miles to the east of the easternmost part of the survey area, also seems unlikely to generate the observed CH<sub>4</sub> surface pattern. The methane concentration in the northeastern, CH<sub>4</sub>-rich part of the survey was increasing with increasing salinity, in contrast to what would be expected from a local freshwater source. In addition, the sharp gradient of the surface CH<sub>4</sub> pattern would be hard to support from a point source more than 30 miles away. Freshwater is, to a large extent, mixed with seawater within Yaquina Bay. Butler *et al.* [1987] showed that the methane carried down by the Yaquina River is almost entirely lost to the atmosphere within the estuary and that the water sampled within the jetty of Newport shows a CH<sub>4</sub> concentration close to the oceanic background throughout the year.

[9] The sea surface temperature (SST) dropped from  $16.5^\circ \text{C}$  in the southwest to  $13.5^\circ \text{C}$  in the east (Figure 2c),

with isotherms in northwest-southeast direction in the eastern part of the survey paralleling the line of a bathymetric high, the Daisy Bank (Figure 2a). Surface temperatures decreased further to  $\sim 9^\circ \text{C}$  closer to the coast, as recorded virtually simultaneously from the R/V *Wecoma* (Figure 3a). The observed upward bending of the isopycnal surfaces (Figure 3a) is caused by summer upwelling, a large-scale phenomenon along the coasts of Oregon and California [Huyer, 1977; Huyer *et al.*, 1998]. The section indicates the presence of upwelled waters at the sea surface during this time. Winds favoring upwelling prevailed on 23–24 July (Figure 4). Surface currents were moderate and equatorward east of  $125^\circ \text{W}$  (Figure 3b). The flow pattern suggests that the observed temperature and CH<sub>4</sub> variations do not result from different surface current regimes.

[10] The increase in CH<sub>4</sub> from southwest to northeast correlates with the drop in SST toward the shelf edge, suggesting a causal link between the enhanced CH<sub>4</sub> concentrations and coastal upwelling (Figure 5). CH<sub>4</sub> values at corresponding temperatures were always higher on the northern transect than on the southern transect of the survey (Figure 5). The CH<sub>4</sub>/SST correlation is also apparent within the whole data set collected in the last half of July 1999 (Figures 6 and 7). However, the CH<sub>4</sub> versus SST relation for the complete data set is more complex, affected by both spatial and temporal variability in the



**Figure 4.** Wind recorded at the wind buoy directly off Newport, at 44.62°N, 124.53°W, at a water depth of ~130 m. A line pointing straight down indicates upwelling-favorable winds from the north. Data have been low-pass filtered to suppress energy with periods shorter than 40 hours. (Source is [http://www.ndbc.noaa.gov/station\\_page.php?station=46050](http://www.ndbc.noaa.gov/station_page.php?station=46050).)

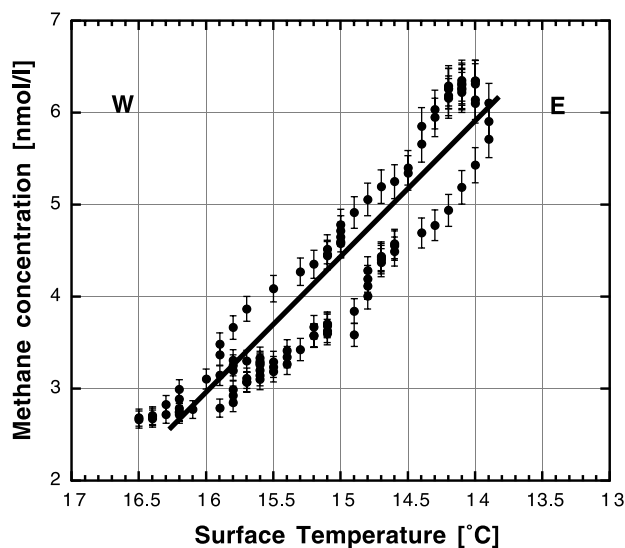
area. Summer upwelling off Oregon is an intermittent process, usually with 4–5 significant upwelling events during the 6-month upwelling season [Huyer, 1976]. A series of short periods of upwelling-favorable winds prevailed during the time of the survey, with highest intensities of northerly winds on 19, 23, and 26 July (Figure 4). Two regions north of the high-resolution survey area were sampled between 125°20'W and 124°55'W during Hydrosweep (HS) surveys HS 38 and HS 47 (for locations, see Figure 6). HS 47 was performed on 23 July, 1 day before the data shown in Figure 2 were sampled. The CH<sub>4</sub>/SST relationships on 23 and 24 July were almost identical (Figure 7), with a slight tendency to higher CH<sub>4</sub> concentrations at given temperatures toward the north, consistent with the north-south gradient observed during the survey on 24 July (Figure 4). On both days, and in both areas, a temperature range of ~2°C (from 16.5° to 14.5°C, from west to east) and a CH<sub>4</sub> concentration range from ~2.7 to 4.5 nmol L<sup>-1</sup> were observed between 125°20'W and 124°55'W. HS 38, covering the area between the high-resolution survey and HS 47 (Figure 6), was performed on 20 July, before the upwelling event on 22–24 July. CH<sub>4</sub> concentrations varied from 3 to 3.5 nmol L<sup>-1</sup>, the longitudinal SST gradient was less prominent (15°–16°C), and the relationship between methane content and surface temperature was less pronounced.

[11] Spatial variability is apparent on close examination of the data gathered during survey HS 26 on 17 July. Although a temperature interval of >2°C (13–15°C) was covered and a reverse correlation between CH<sub>4</sub> content and SST was observed, the slope of the CH<sub>4</sub>/SST plot is insignificant. This seems to indicate a lower content of the upwelled source waters at that time and place. However, different velocities for heat (surface temperature) and CH<sub>4</sub> transfer could also be an explanation for the less pronounced temperature dependence of the CH<sub>4</sub> content. To estimate the timescale for the ventilation of the mixed layer, we used the equation  $k = 0.39u^2(Sc/660)^{-1/2}$  suggested by Wanninkhof [1992], where  $k$  is the piston velocity,  $u$  is the long-term average wind speed, and  $Sc$  is the Schmidt number for methane at the ambient conditions. With a mean wind speed of 4.8 m s<sup>-1</sup> and a Schmidt number of 860 at the mean

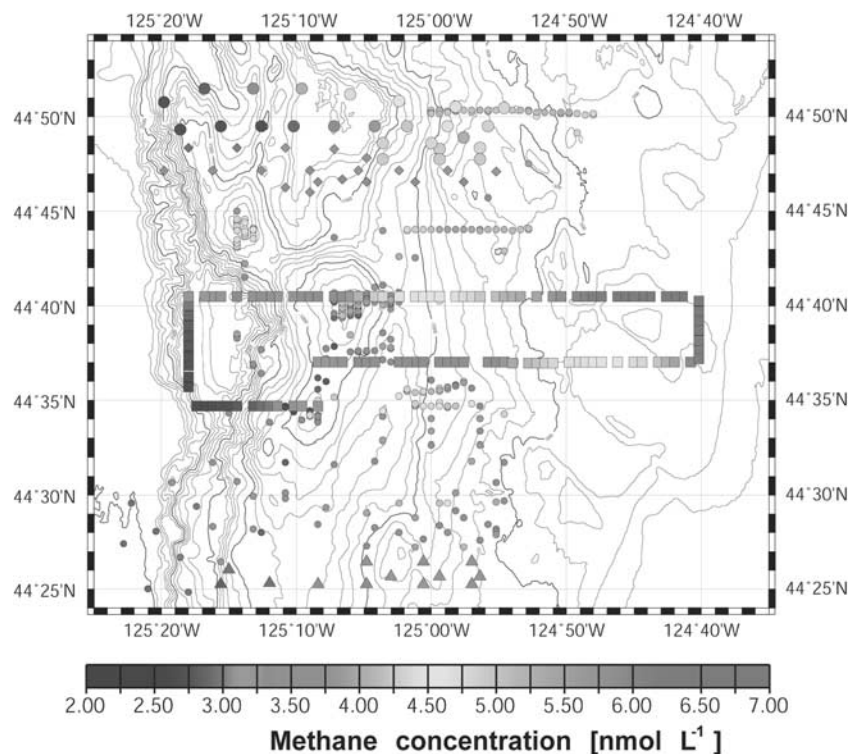
temperature of 15°C, the piston velocity is ~8 cm h<sup>-1</sup> or 2 m day<sup>-1</sup>. The mixed layer depth throughout the survey was between 9 and 14 m. A correlation of the mixed layer depth and the wind stress pattern during the survey was not observed (data not shown). Under these conditions the timescale for a water mass newly brought to the surface to lose its excess methane content to the atmosphere should be slightly shorter than a week. Hence the methane concentration in the waters surveyed is dependent not only on the CH<sub>4</sub> content of the upwelled waters but also on the time elapsed between the last upwelling event and the time of the measurement.

[12] Waters brought to the surface by upwelling should also be characterized by a higher salinity. As pointed out earlier, the lowest methane contents during the survey on 24 July (Figure 2) were found in the waters with lowest salinities in the southwestern edge of the survey area; the highest CH<sub>4</sub> concentrations in the northeastern corner coincided with comparatively high salinities ( $S > 31$ ). However, there is no good correlation for the entire data set (Figure 2d). These findings can be explained by mixing of upwelled water (high CH<sub>4</sub>, low SST, and high  $S$ ) with surface water that is considerably warmer but has a strong variation in surface salinity due to spatial variability of the influence of the Columbia River plume and precipitation [van Geen et al., 2000].

[13] We will discuss two possible processes that would establish the relationship between coastal upwelling and high methane concentrations in surface waters: (1) enhanced in situ production of methane driven by high primary production, abundance of particles, and zooplankton activity, and



**Figure 5.** Correlation of CH<sub>4</sub> content and surface water temperature from survey illustrated in Figures 2b and 2c. CH<sub>4</sub> concentrations at corresponding temperatures were higher on the northern east-west transect than on the southern east-west transect, suggesting that the upwelled waters have higher concentration toward the north. Note that the  $x$  axis is reversed for compatibility with a view from west to east.



**Figure 6.** Map showing positions of all CH<sub>4</sub> surface concentration measurements made in July 1999 (except transfers to harbors). Absolute concentrations are indicated by symbol color, using the same color scale as that used for Figure 2c. Locations of parts of the data set discussed in text are indicated by different symbols: squares, high-resolution survey on 24 July; large dots, Hydrosweep (HS) 47 on 23 July; diamonds, HS 38 on 20 July; triangles, HS 26 on 17 July; small dots, all other data. See color version of this figure at back of this issue.

(2) advective transport of methane-rich subsurface waters to the surface. As discussed below, our data strongly suggest the subsurface source.

[14] Slightly elevated concentrations of methane have been reported as a result of enhanced biological production of CH<sub>4</sub> in equatorial upwelling regimes [Bates *et al.*, 1996]. In situ production of methane in the upper water column within anaerobic microenvironments provided by zooplankton or fecal pellets was shown to produce subsurface maxima directly below the mixed layer. This vertical methane distribution is a result of the combined effects of decreasing production rates with depth and the enhanced loss to the atmosphere in the mixed layer by air-sea exchange [Karl and Tilbrook, 1994; Tilbrook and Karl, 1995].

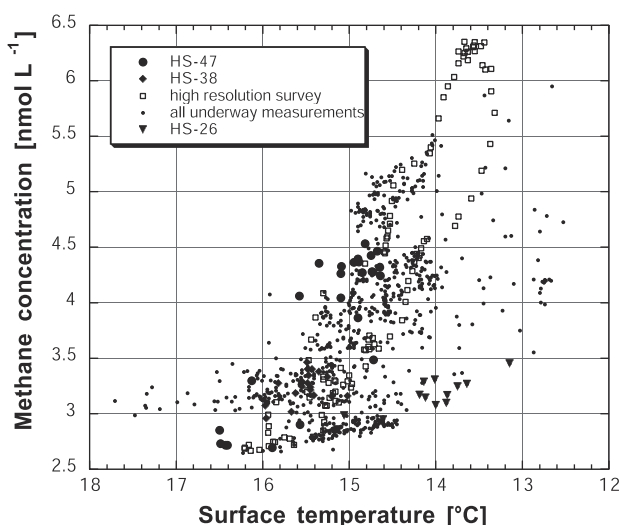
[15] In situ production has also been suggested to cause enhanced CH<sub>4</sub> fluxes in areas of coastal upwelling in connection with suboxic or anoxic conditions at shallow water depth, as in the Arabian Sea [Bange *et al.*, 1998; Owens *et al.*, 1991] or, more recently, in the eastern tropical North Pacific [Sansone *et al.*, 2001]. In these cases, high primary production due to upwelling leads to anoxic conditions at water depths of <200 m. High CH<sub>4</sub> concentration has been found at the oxic/anoxic interface

and has been suggested to be caused by the anoxic degradation of organic matter at these depth levels [Owens *et al.*, 1991]. The methane produced at these depths can be transported actively to the surface by upwelling. Bange *et al.* [1998] showed a negative correlation between SST and methane concentration for surface waters in the northwestern Arabian Sea, which is similar to our findings off Oregon.

[16] Coastal upwelling has been discussed as a mechanism to enhance CH<sub>4</sub> fluxes to the atmosphere in the California Current system for the coast off southern California [Cynar and Yayanos, 1992, 1993]. The methane distribution between Point Conception and San Diego in November 1989 and March 1990 revealed a higher methane inventory in the upper 300 m of the water column as well as a higher flux to the atmosphere during upwelling in March of 1990. The hydrographic parameters indicated moderate upwelling in March 1990 but indicated no upwelling in November 1989. The authors suggested that upwelling enhances the net air-sea flux of methane both through transport of methane-rich subsurface waters to the surface and through enhanced in situ production.

[17] However, enhanced in situ production of methane is not consistent with the observed pattern off Newport. No





**Figure 7.** CH<sub>4</sub> concentration versus sea surface temperature for entire survey. Different parts of data set discussed in text are indicated by different symbols: squares, high-resolution survey on 24 July; large points, data collected during HS 47 on 23 July; diamonds, data collected during HS 38 on 20 July; triangles, data collected during HS 26 on 17 July; small dots, all other CH<sub>4</sub> surface data. Estimated error for CH<sub>4</sub> measurements is <2%, except for the manual high-resolution survey, where estimated error is 3.5%.

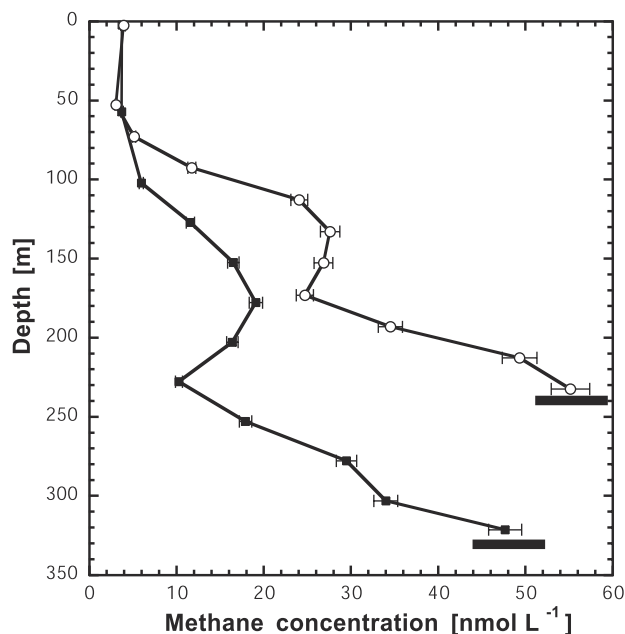
pronounced subsurface maxima were observed below the mixed layer at the stations off Newport either in summer or in fall (see Figures 8 and 9). The water column is not anoxic at any depth level, and even suboxic conditions are encountered only below 500 m [Torres *et al.*, 1998], which is well below the source depth of the upwelled waters in this region [Barber and Smith, 1981; van Geen *et al.*, 2000]. In addition, the correlation of CH<sub>4</sub> versus SST falls on two different lines for the northern and the southern across-slope transects (Figure 5). At a given SST, methane concentrations were always higher on the northern leg of the survey. This suggests a strong spatial gradient that would be difficult to explain with variations of in situ production in the same upwelling controlled hydrographic setting.

[18] The high CH<sub>4</sub> concentrations at the surface could also be caused by the upwelling of waters enriched in CH<sub>4</sub> derived from seafloor sources, for example, seepage. Bottom sources for methane have been located at various depths in or near the survey area. Focused venting of methane-rich fluids has been observed at water depths of ~2100 m on the northwestern edge of the Western Basin (Figure 1, ~44°41'N, 125°W [Carson *et al.*, 1990]). Seepage of methane-bearing fluids and ebullition of methane gas have been reported at Hydrate Ridge (water depth of 600–800 m). This source has been shown to generate CH<sub>4</sub> plumes up to 200 m above the sediment surface, i.e., up to <400 m water depth [Torres *et al.*, 1998; Collier *et al.*, 1999]. The fluid and gas expulsions at Hydrate Ridge have recently been shown to be enforced by the decomposition of gas hydrates [Suess *et al.*, 1999].

[19] Active seepage on the shelf leads to strong, local enrichment in the lower water column both to the north

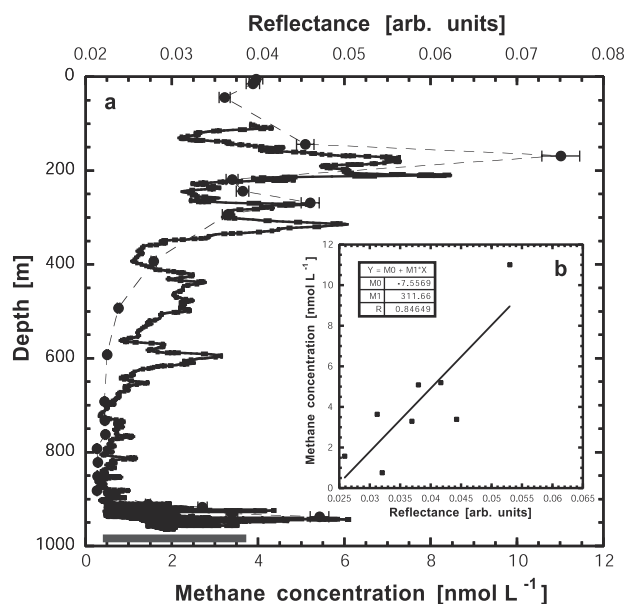
and to the south of the survey area [Collier and Lilley, 1995; Lilley *et al.*, 1990]. Methane concentration profiles from stations in the vicinity of an active gas seepage site on the Oregon shelf near 43°01.84'N, 124°40.16'W (Coquille Bank, near Coos Bay) showed CH<sub>4</sub> concentrations of >150 nmol L<sup>-1</sup> close to the bottom. The isotopic composition and C<sub>1</sub>/C<sub>n</sub> ratio suggest a thermogenic CH<sub>4</sub> source. A gas seep with similar isotopic characteristics has been found at Hecata Bank (44°00.2'N, 124°52.2'W). Both seeps are located at depths shallower than 200 m.

[20] Increasing CH<sub>4</sub> concentrations of up to 60 nmol L<sup>-1</sup> in bottom waters near the upper shelf break have also been found along the southern leg of the survey area in October 1999 (Figure 8). Both stations show enhanced methane concentrations toward the bottom. However, the surface water saturation stayed well below 130% (3.7 nmol L<sup>-1</sup>), significantly lower than during summer upwelling at the same site. The profiles show a minimum at 50 m depth, which indicates that the high CH<sub>4</sub> content of the subsurface and bottom waters had minor impact on the surface water CH<sub>4</sub> concentration during this time. However, when advected to the surface during coastal upwelling in summer, these waters would be strongly oversaturated with respect to the atmosphere. The high CH<sub>4</sub> inventory below the surface in autumn, in combination with the distinctly higher CH<sub>4</sub> surface concentrations during a summer upwelling event, documents the importance of upwelling of methane



**Figure 8.** Methane profiles from Station 5 (squares) and Station 6 (circles; for location see Figure 1) sampled in October 1999 on R/V *Wecoma*. Black bars indicate seafloor depth. Both stations show enhanced methane concentrations toward the bottom. However, surface water saturation stays well below 130%, significantly lower than during summer upwelling at the same location. Concentration minima at 50 m depth indicate that high CH<sub>4</sub> content of subsurface and bottom waters has only minor impact on release of methane to the atmosphere in autumn.





**Figure 9.** (a) Methane profile (points, dashed line) at Station 67 of R/V *Sonne* cruise 143-1b on the northwest knoll and response of a reflectance sensor installed on the Oregon State University zero angle photon spectrometer (ZAPS) system [Klinkhammer, 1994] deployed at the same station 8 hours earlier (solid line, in arbitrary units). Shaded bar indicates water depth. CH<sub>4</sub> maximum between 150 and 300 m is related to higher abundance of reflecting particles. Increase of CH<sub>4</sub> toward bottom is related to fluid flow on the northwest knoll as indicated by observation of bacterial mats, Calyptogena, etc. [Bohrmann et al., 2000]. (b) Correlation of methane concentration and reflectance in depth range from 100 to 400 m.

dissolved at greater depths to overcome the stratification barrier.

[21] The Daisy Bank, in the northeastern part of the high-resolution survey area, is bounded in the south by the Daisy Bank Fault, a major active strike-slip fault. Carbonate structures (chimneys, doughnuts, and slabs) indicating fluid flow in the past have been found to be abundant in close proximity to the fault [Goldfinger et al., 1996]. Active gas venting was observed on some of the strike-slip faults on the Cascadia margin during dives with the submersible Delta in 1992 and 1993 (C. Goldfinger, personal communication, 2000). The presence of gas seeps at the southwestern flank of the Daisy Bank in late summer 1999 has been inferred from a 4-kHz Parasound record, during cruise 143/3 of R/V *Sonne* [Bohrmann et al., 2000]. The record shows the occurrence of a 3-km-wide zone of weak reflectivity in the water column, which was restricted to a narrow band between 345 and 370 m water depth, never more than 30 m above the seafloor. However, response in the water column to the Parasound signal could be caused by other phenomena (i.e., fish swarms or particle layers). A dive with the Canadian remote operated vehicle ROPOS deployed from R/V *Sonne* in summer 2000 revealed layers of fine suspended material but revealed

no gas ebullition at the site (G. Bohrmann, personal communication, 2000). Hence it remains unclear whether there is recent active gas venting in the immediate vicinity of the Daisy Bank.

[22] We commonly observed high methane concentrations in well-defined layers within the depth range of 150–400 m that were correlated with enhanced backscatter and reduced light transmission (Figure 9; see also data from Torres et al. [1998]). Off northern California the cross-shelf transport of particle-rich layers (intermediate nepheloid layer (INL)) has been shown to be driven by the occurrence of mesoscale eddies [Washburn et al., 1993]. Organic-rich resuspended material from the seafloor on the shelf and on the shelf break was transported offshore for several tens of kilometers. Both mesoscale eddies and the occurrence of INLs are common features along the California Current system and have been observed as far to the north as the survey area [Pak and Zaneveld, 1978; Pak et al., 1980b; Barth et al., 2000]. A weak correlation of suspended particle abundance and methane concentration has been reported for the suboxic waters of the Arabian Sea [Jayakumar et al., 2001], but the correlation was not found to be one-to-one in all cases, and maxima in beam attenuation and CH<sub>4</sub> concentration did not always coincide. Burke et al. [1983] have investigated the correlation of methane and suspended matter in a section along the west coast of Central America between 25°N and 5°N. The authors found no statistically relevant correlation between the two parameters for the entire section. However, the two northernmost stations, which are the only stations of the survey within the California Current system, show a strong correlation of methane and particle concentration. Our observations off Oregon are in general agreement with the observations at the stations in the California Current. Whether the high methane concentrations in connection with nepheloid layers is a local feature off Oregon (and off Baja, California [Burke et al., 1983]) needs further investigation. At this point, it also remains unclear whether the enhanced CH<sub>4</sub> concentrations in the plume are caused by in situ production on particles or whether the particle-containing waters already bear enhanced methane levels where they detach from the shelf after interaction with reduced sediment and enclosed pore waters. In any case, the process results in the formation of methane-rich shallow layers, which could be transported upward by coastal upwelling.

[23] In summary, the data strongly suggest that the increasing oversaturation toward the coast observed off Oregon in summer is related to the upwelling of subsurface waters, which are enriched in CH<sub>4</sub> from several possible bottom sources. Whether venting from the well-characterized gas sources at Hydrate Ridge can affect newly upwelled surface waters seems questionable. However, the observation of gas plumes, indicated by acoustic anomaly patterns to rise at least 200 m above the summit of Hydrate Ridge, shows that gas may penetrate to water depths as shallow as 400 m. The source waters for upwelling off Oregon generally originate from depths between 100 and 200 m [Barber and Smith, 1981]. Recently, the source depth for surf-zone waters was estimated to be 100–150 m in August 1995, based on the distribution of nutrients [van

Geen et al., 2000]. However, higher surface nutrient concentrations observed in August 1996 imply a source depth of 250–320 m [Takesue and van Geen, 2002]. Thus it is very likely that methane from sources on the shelf, as well as from beyond the shelf break and upper slope (<300 m), may be brought to the surface and may be subject to exchange with the atmosphere.

#### 4. Significance

[24] The enhancement of CH<sub>4</sub> fluxes to the atmosphere in regions of coastal upwelling is likely to occur on a global scale. Coastal upwelling is a first-order phenomenon along large parts of the global continental margins [Barber and Smith, 1981; Suess and Thiede, 1983]. It can increase fluxes to the atmosphere considerably through enhanced in situ production, in particular in areas of shallow anoxia, combined with a pathway for advective upward methane transport [Bange et al., 1998; Burke et al., 1983; Sansone et al., 2001]. Apart from this direct link, there is a strong coincidence between regions of coastal upwelling and the occurrence of methane sources from the seafloor. Oil and gas seepage, groundwater discharge, and marine gas hydrates are exclusively found along continental margins [Hovland and Judd, 1988; Kvenvolden, 1998]. The onset of methanogenesis, leading to gassy sediments, to the formation of reservoirs of biogenic methane, and, under suitable pressure-temperature conditions, to the formation of gas hydrates, is restricted to areas with high export of organic matter to the seafloor. The areas in which the concentration of organic carbon is larger than 0.5%, which is often considered the minimum value for the formation of gas hydrates [Gornitz and Fung, 1994; Harvey and Huang, 1995], are almost entirely restricted to the ocean margins (upper slope and shelves) and the marginal seas [Premuzik et al., 1981]. The high primary production in coastal upwelling areas leads to particularly high abundance of organic carbon and hence to conditions favoring methane production in sediments.

[25] Intermediate nepheloid layers that detach from the shelf and upper slope and transport particles offshore have been reported in several upwelling areas [Kullenberg, 1981; Pak et al., 1980a; Pak and Zaneveld, 1978; Pak et al., 1980b; Washburn et al., 1993; Jayakumar et al., 2001; Burke et al., 1983] at water depths between 100 and 400 m, including parts of the west coasts of North America, South America, Africa, and the Arabian Sea. Evidence for high concentrations of dissolved CH<sub>4</sub> within these layers has been shown for selected stations [Jayakumar et al., 2001; Burke et al., 1983] and is now evident for the southern and northern end of the California Current system (this work and that of Burke et al. [1983]). It remains to be shown that INLs in other upwelling regions are also associated with a high CH<sub>4</sub> content.

[26] The upward advective transport of water in coastal upwelling regions appears to be an important process for the understanding of global scenarios affecting the decomposition of methane hydrates. Such scenarios, explaining carbon isotopic excursions in the past [Kennett et al., 2000; Dickens et al., 1997, 1995] as well as assessing the importance

of gas hydrates in the present framework of global warming, have so far neglected the interaction with the ocean. As shown by Harvey and Huang [1995], it is of great significance whether methane from bottom sources enters the atmosphere in the form of CH<sub>4</sub> or of its oxidation product, CO<sub>2</sub>. Coastal upwelling provides a mechanism for transporting methane to the ocean surface, shortening the time that the gas resides in the water column and is subject to microbial oxidation, thus amplifying methane emission to the atmosphere to a degree that is, at present, entirely unknown.

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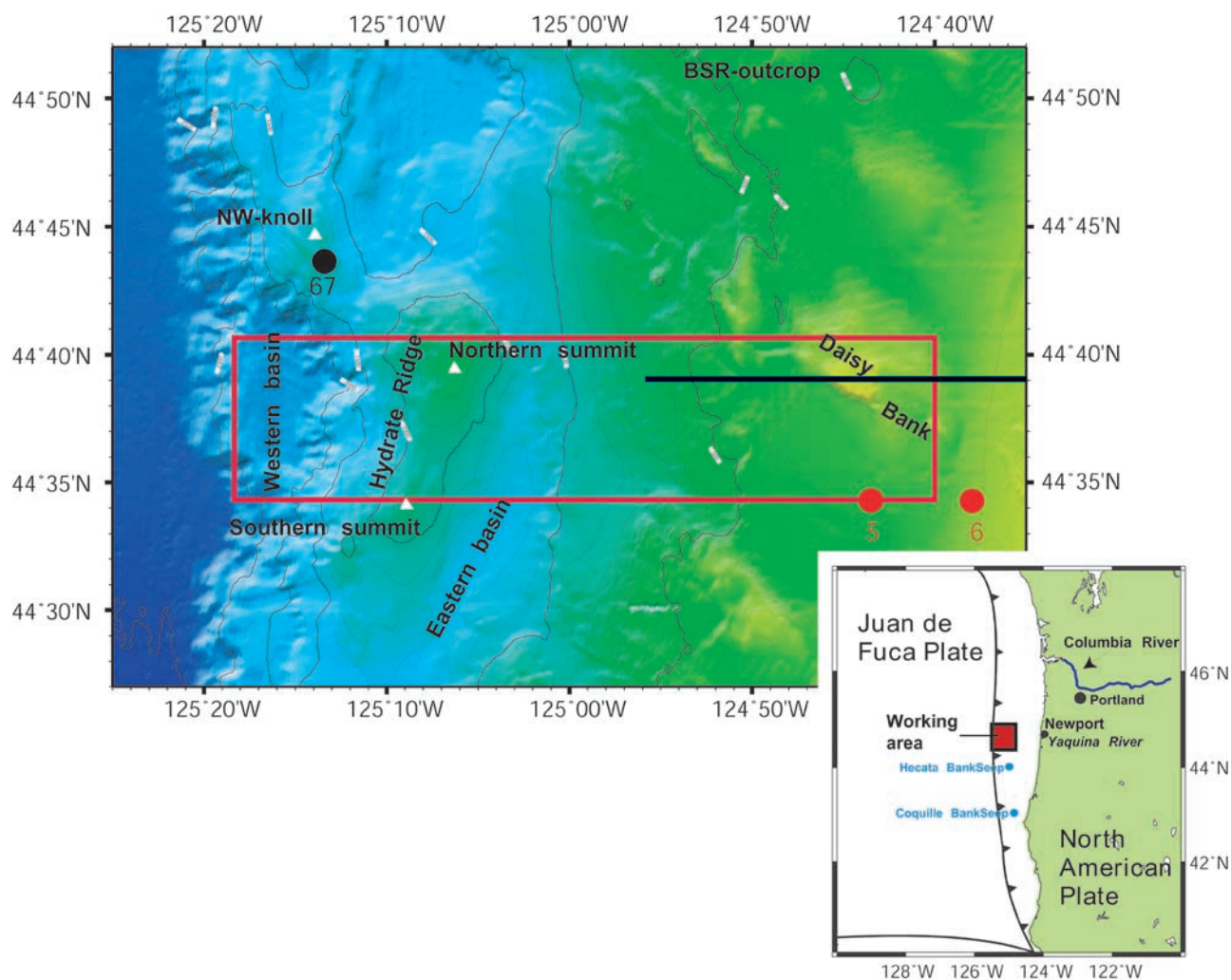
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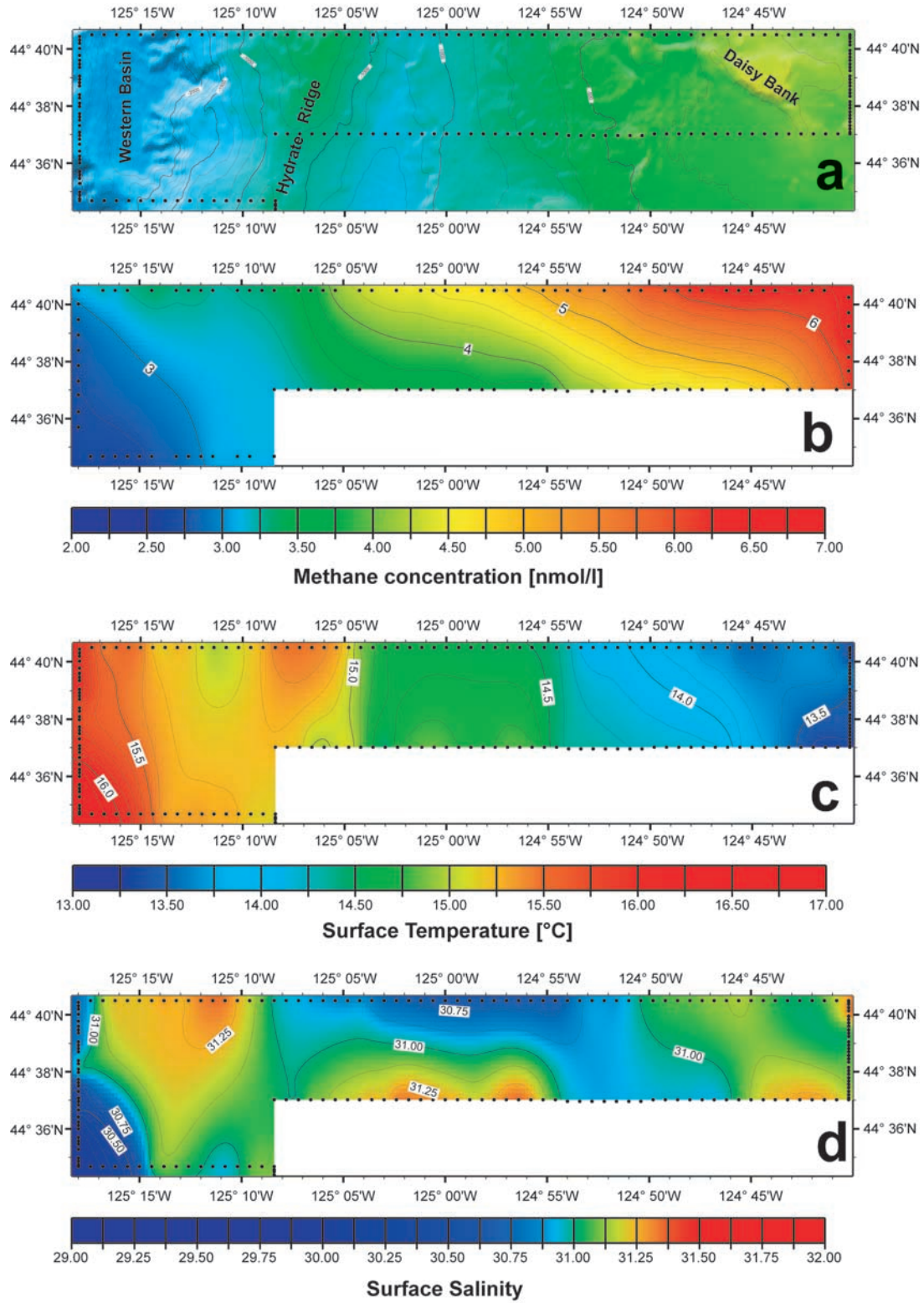
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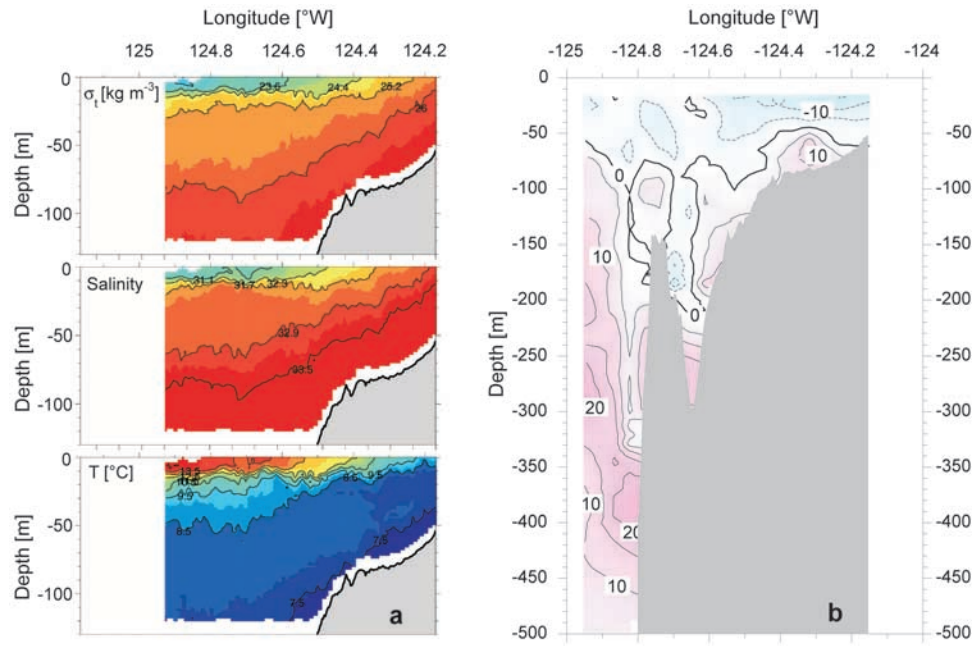




**Figure 1.** Map of working area on the continental margin off Oregon. Red rectangle denotes region of surface survey from 24 July shown in Figures 2a–2d. Black line is western part of track of current, temperature, and salinity section from same day shown in Figures 3a and 3b. Red dots indicate hydrocast stations from October 1999, shown in Figure 8. Black dot shows location of Station 67, chosen to illustrate occurrence of midwater CH<sub>4</sub> maxima in connection with intermediate nepheloid layers (Figure 9). More locations specified in the text, as well as the subduction front, are indicated in overview map (lower right).

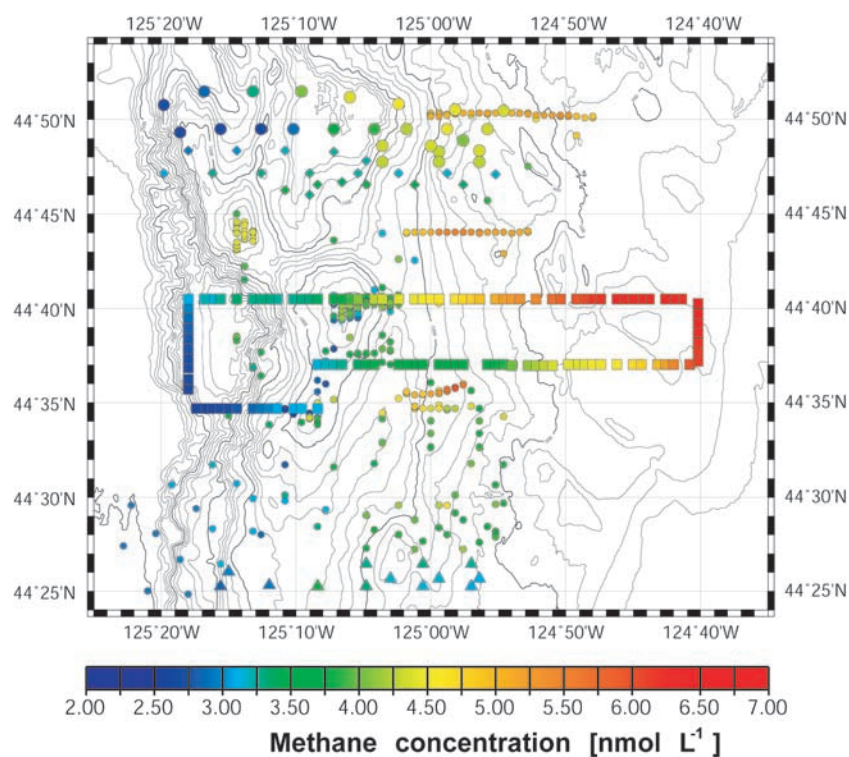


**Figure 2.** Results from a high-resolution CH<sub>4</sub> surface survey from 24 July 1999 showing (a) bathymetry; (b) surface methane concentration, (c) surface seawater temperature, and (d) surface seawater salinity. Black dots indicate locations of individual measurements.



**Figure 3.** (a) The  $\sigma_t$ , salinity, and temperature sections from a SeaSoar survey and (b) section of alongshore current (in cm s<sup>-1</sup>, positive northward) derived from an acoustic Doppler current profiler survey. Both data sets along 44°39'N off Newport were recorded on 24 July 1999 on R/V *Wecoma* (see Figure 1 for location, but note that the section extends 30 km farther east than shown in Figure 1). Flow pattern suggests that observed temperature and CH<sub>4</sub> variations do not result from different surface current regimes. *SeaSoar* data indicate the presence of upwelled waters at the sea surface during that time.





**Figure 6.** Map showing positions of all CH<sub>4</sub> surface concentration measurements made in July 1999 (except transfers to harbors). Absolute concentrations are indicated by symbol color, using the same color scale as that used for Figure 2c. Locations of parts of the data set discussed in text are indicated by different symbols: squares, high-resolution survey on 24 July; large dots, Hydrosweep (HS) 47 on 23 July; diamonds, HS 38 on 20 July; triangles, HS 26 on 17 July; small dots, all other data.